



# Temporal variations and trends of CFC11 and CFC12 surface-water saturations in Antarctic marginal seas: Results of a regional ocean circulation model

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## ABSTRACT

The knowledge of chlorofluorocarbon (CFC11, CFC12) concentrations in ocean surface waters is a prerequisite for deriving formation rates of, and water mass ages in, deep and bottom waters on the basis of CFC data. In the Antarctic coastal region, surface-layer data are sparse in time and space, primarily due to the limited accessibility of the region. To help filling this gap, we carried out CFC simulations using a regional ocean general circulation model (OGCM) for the Southern Ocean, which includes the ocean–ice shelf interaction. The simulated surface layer saturations, i.e. the actual surface concentrations relative to solubility-equilibrium values, are verified against available observations. The CFC surface saturations driven by concentration gradients between atmosphere and ocean are controlled mainly by the sea ice cover, sea surface temperature, and salinity. However, no uniform explanation exists for the controlling mechanisms. Here, we present simulated long-term trends and seasonal variations of surface-layer saturation at Southern Ocean deep and bottom water formation sites and other key regions, and we discuss differences between these regions. The amplitudes of the seasonal saturation cycle vary from 22% to 66% and their long-term trends range from 0.1%/year to 0.9%/year. The seasonal surface saturation maximum lags the ice cover minimum by two months. By utilizing observed bottle data the full seasonal CFC saturation cycle can be determined offering the possibility to predict long-term trends in the future. We show that ignoring the trends and using instead the saturations actually observed can lead to systematic errors in deduced inventory-based formation rates by up to 10% and suggest an erroneous decline with time.

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## 1. Introduction

Ocean tracer observations are a powerful tool to understand the formation and spreading of water masses. Sampling for the chlorofluorocarbons CFC11 and CFC12 has become a routine on many physical oceanography cruises, resulting in a large number of observations, in particular since the WOCE period (Orsi et al., 2002). Often, only tracer measurements highlight the spreading paths of freshly ventilated water masses, which stand out as spatially limited regions with tracer concentrations higher than in the surrounding waters.

Compared to the “classical” tracers, like oxygen and nutrients, anthropogenic CFC11 and CFC12 have the advantage to be chemically

inert. CFCs enter the ocean by gas exchange across the ocean–atmosphere interface. Since their first appearance in the 1930s and up to around the turn of the millennium (Walker et al., 2000), the atmospheric concentrations increased (Fig. 13a), which is reflected in the temporal evolution of their concentrations in the ocean. This transient signal allows one to derive for tracer-carrying water masses:

- tracer ages, for example, Weiss et al. (1985), Thiele and Sarmiento (1990), Doney et al. (1997),
- age/transit time distributions (TTD), for example, Beining and Roether (1996), Hall et al. (2002), Waugh et al. (2002, 2003), Huhn et al. (2008b),
- formation and subduction rates, for example, Broecker et al. (1999), Orsi et al. (1999), Meredith et al. (2001), Smethie and Fine (2001), Rhein et al. (2002), Kieke et al. (2006).

Table 1 lists observed mixed layer saturations in high latitudes, where the CFC saturation is defined as the ratio of the actual concentration to the concentration in equilibrium with the

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**Table 1**  
Observed CFC saturations in the ocean mixed layer.

Region year	Saturation (%)	Source	Abbr.
<i>Shelf water mass at Antarctic's Periphery</i>			
Unspecified	40–60	Orsi et al. (2001)	
<i>Ross Sea</i>			
1984	CFC11: 64	Trumbore et al. (1991) and	R:1
1984	CFC12: 79	Smethie and Jacobs (2005)	
1994	CFC11: 74	Smethie and Jacobs (2005)	R:2
1994	CFC12: 79		
1994	60–70/90	Orsi et al. (2002)	R:3
2000	CFC11: 84	Smethie and Jacobs (2005)	R:4
2000	CFC12: 90		
2000/2001	72–95	Rivarolo et al. (2004)	R:5
<i>Wilkes Land</i>			
1994	70	Orsi et al. (2002)	
<i>Amery Ice Shelf, Prydz Bay</i>			
1994	70	Orsi et al. (2002) and Haine et al. (1998)	A:1
<i>Central Weddell Sea</i>			
1987	90	Mensch et al. (1996)	W:2
1992	65	Sültenfuß (1998)	
1996	85	Sültenfuß (1998)	W:3
<i>Western Weddell Sea</i>			
1992	55–85	Mensch et al. (2000)	W:1
1996	65	Sültenfuß (1998)	W:4
2004	68–70	Huhn et al. (2008a)	W:5
<i>Southern Weddell Sea, in front of the Filchner Ice Shelf</i>			
1985	85	Mensch et al. (1996)	F:1
1987	65	Mensch et al. (1996)	F:2
1987	70–75	Schlosser et al. (1991)	F:3
1993	85–90	Gammelsrød et al. (1994)	F:4
<i>Labrador Sea (Northern hemisphere)</i>			
1986	60	Wallace and Lazier (1988)	
1988–1992	60–70	Smethie et al. (2000) and Smethie and Fine (2001)	
1988–1992	66	Terenzi et al. (2007)	
1997	CFC11: 90 ± 8	Azetsu-Scott et al. (2005)	
1997	CFC12: 95 ± 9		

The abbreviations of the last column are for reference further below. Separately reported values for CFC11 and CFC12 are indicated.

atmospheric CFC partial pressure. For deeper waters, similarly, one uses the term “apparent saturation”, the reference concentration being the concurrent equilibrium concentration at the ocean surface at the observed temperature and salinity. This saturation is termed apparent, since it ignores the fact that, when the waters actually descended from the mixed layer, the equilibrium concentration was lower than the concurrent one. Consequently, the apparent saturation is lower than the actual.

On the microscopic scale, CFC uptake occurs by diffusion through the oceanic skin layer on top of the mixed layer. The process is rather slow (equilibration time scale for a 100-m deep mixed layer is on the order of one month) so that the actual CFC uptake is controlled by mixed layer processes, essentially the exchange between the mixed layer and deeper layers (mixed layer entrainment/detrainment). The resulting undersaturation (Table 1) defines the air–water CFC gradient, to which the CFC uptake is proportional. Consequently, the sequestration of fluid in the seasonal thermocline and variations in the depth of mixing are primary factors (Haine and Richards, 1995), which need to be adequately represented for a realistic simulation of tracer concentrations and inventories in deep and bottom waters (Doney and Jenkins, 1988). Vertical mixing and other dynamical factors in the upper ocean are also essential for a realistic simulation of the Southern Ocean's hydrographic structure and sea ice coverage (Timmermann and Beckmann, 2004).

In the Southern Ocean, the interaction with sea ice, the ice shelves, and the atmosphere transforms local water masses into deep and bottom waters. On broad continental shelves, the accumulation of High Salinity Shelf Water (HSSW), due to brine

release by sea ice formation, initiates two known mixing processes, namely:

- The Foldvik- or ISW-process (Foldvik et al., 1985): HSSW flows into ice shelf caverns where it is modified to Ice Shelf Water (ISW) due to the interaction with the ice shelf base.
- The Foster and Carmack (1976) process: Locally formed HSSW mixes with pycnocline waters penetrating onto the continental shelf.

Both mixing products descend along the continental slope under entrainment of ambient water masses to form deep or bottom water, depending on the entrainment rate.

During the transition from winter to summer, melting sea ice forms a seasonal halocline at 20–50 m depth (Carmack, 1974). The Antarctic Surface Water (ASW) above has temperatures and salinities ranging from –1.8 to 2.0 °C and from 33.0 to 34.3, respectively. The deeper layer, called Winter Water (WW) (Carmack, 1974), maintains the characteristics of the Winter Mixed Layer (WML) with temperatures near the surface freezing point.

Since the beginning of CFC observations in the Southern Ocean marginal seas in 1985 (Mensch et al., 1996), observations, which have primarily been collected during the austral summer, remain sparse in space and time. Therefore, it has been common to assume a time-invariant saturation on the basis of the available data (Table 1).

Existing CFC observations in surface waters are far from resolving the seasonal to interannual variability (Section 3). Thus,

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